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Title: Progress towards waterproofing uranium nitride

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Introduction

Uranium mononitride (UN) is a promising, high-uranium-density fuel because of its good thermal properties and performance during irradiation and has been considered as a potential replacement for uranium(IV) oxide (UO_2) in commercial light water reactors. Despite these advantages, limitations are observed when the coolant comes in contact with the fuel, e.g. breach of cladding, resulting in degradation of the fuel. Research focused on continuing work from FY18 that examined the feasibility of UN-UO_2 composites for UN waterproofing purposes. Steam oxidation testing of candidate waterproofing additive materials was performed to evaluate the corrosion resistance of these materials. Composite pellets were also pressed and heated to examine chemical compatibility with UN.

Project description

Previous work in FY18 evaluated UN and UN-UO_2 composites for resistance to oxidation in steam. During isothermal oxidation tests, mass gain and pulverization occurred over the course of minutes, though the addition of UO_2 significantly delayed the onset of oxidation. Similarly, during temperature ramps in steam, the addition of UO_2 appeared to delay the onset of oxidation, though pulverization occurred for pellets containing more than 10 volume percent uranium mononitride. This work showed that waterproofing effects are noticeable with compositing with oxidation-resistant mateirals.

In FY19, compositing efforts focused on evaluating UN cermets with metals that exhibit resistance to steam corrosion, such as chromium (Cr), silicon (Si), and yttrium (Y). UN-SiC composites were also examined for feasibility. Materials tested for steam oxidation were assessed for corrosion resistance based on the percentage of the material oxidized (a reaction coordinate) in steam. Chemical compatibility between UN and additive materials was also assessed using X-ray diffraction after heating.

Accomplishments

The goal of the steam oxidation experiments was to determine the oxidation resistance of the candidate additive materials for compositing with UN, while the goal of the chemical compatibility testing was to evaluate chemical interactions between UN and the candidate additive materials.

Steam oxidation testing was performed on samples with known masses in temperatures ranging between 200 and 1000 °C using a steam furnace coupled with thermogravimetric analysis at the Fuels Research Laboratory at LANL. Samples were heated in argon gas maintained at oxygen levels below 0.1 ppm; once the desired temperature was reached, the steam/argon mixture was flowed through the system and sample mass change as a function of temperature and time was recorded. Results of the steam oxidation testing are shown in [figure 1]. Results showed that, aside from sponge zirconium, all the materials exhibited adherent oxide layers. It was observed that chromium, silicon, APMT, and silicon carbide exhibited the best resistance to corrosion (little-to-no mass gain). Based on these results, chemical compatibility testing was performed between UN and Y, Cr, Si, and SiC.

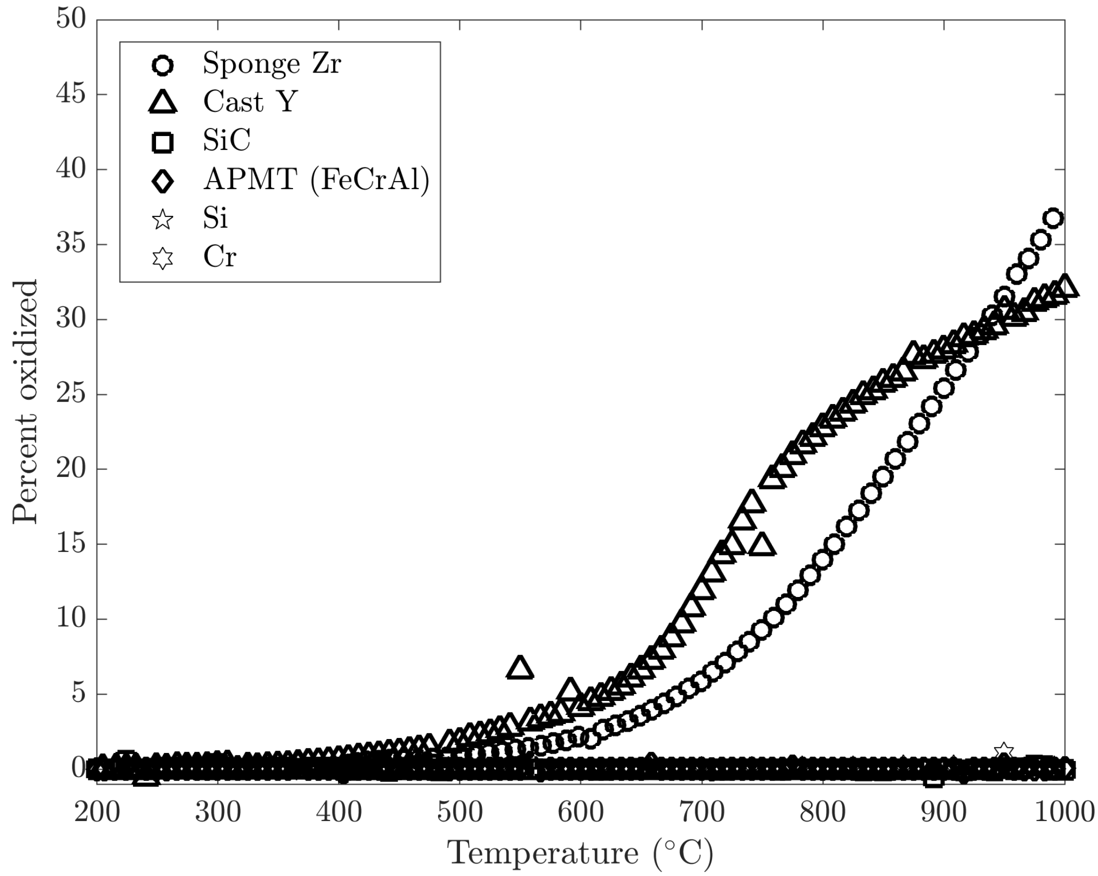

Figure 1: Oxidation fraction, in percentage, as a function of temperature for various candidate additives for UN waterproofing applications in steam up to 1000 °C. It was found that SiC, APMT, Si, and Cr exhibited the best resistance to steam corrosion.

Composite pellets of UN with Y, Cr, Si, and SiC were fabricated by mixing powders of the aforementioned materials. Yttrium dihydride powder was used in lieu of yttrium metal powder due to improved ability to mechanically mill the material. Composites with Y, Cr, and Si were heated to temperatures above the melting points of the respective metals (1526, 1857, and 1410 °C) with the aim of evaluating chemical interactions during liquid-phase sintering. Composites with SiC were heated under temperature profiles used for two-step, pressure-less sintering of SiC.

Results showed that yttrium absorbed nitrogen from the uranium, resulting in the formation of yttrium mononitride (YN) and uranium metal. Conversely, silicon and silicon carbide appeared to react with the material. In the case of silicon, the resultant pellets comprised of primarily uranium silicides. It is not yet clear what reactions occurred with silicon carbide, but the system is being studied further due to interest in such composites. The composites with chromium showed little-to-no remaining chromium metal due to the high vapor pressure of chromium at the temperatures required for liquid-phase sintering. As a result of these various outcomes, the FRL has developed several selection criteria for future waterproofing concepts. Future concepts should be evaluated on the basis of reactivity with uranium (should not form stoichiometric compounds), reactivity with nitrogen (should not absorb nitrogen from uranium), vapor pressure (should not significantly volatilize), and resistance to waterside corrosion (should form a protective oxide layer over a wide temperature range). Finally, cermet and ceramic composite concepts should also preferentially oxidize with respect to uranium mononitride so as to prevent fuel pulverization.

Publications
N/A

Write one sentence on why this project is important
This project has made significant advances in understanding the difficulties that arise from nuclear fuel waterproofing efforts and has resulted in selection criteria that will be useful in evaluating future waterproofing concepts.